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Probing Phase Transitions in Ultrathin Ferroelectric Films Using *In Situ* X-ray Scattering

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Ionic or electronic charge compensation at the surface of an ultrathin ferroelectric film is critical in determining the polarization structure of the complete film. Grazing-incidence x-ray scattering (GIXS) provides a powerful probe of the atomic displacements that underlie this polarization structure, enabling tracking of phase changes and domain formation in ferroelectric films. Performing GIXS measurements *in situ* allows us to study the roles that temperature and chemical environment play in determining the equilibrium polarization of a ferroelectric film. We present here studies of the equilibrium polarization in epitaxially strained $PbTiO_3$ films on conducting $SrRuO_3$ electrodes on (001) $SrTiO_3$ as a function of temperature, chemical environment, and film thickness. Changes in the ionic compensation at the surface due to changes in the oxygen partial pressure of the environment produce strong effects on the Curie temperature. High or low oxygen partial pressures (pO_2) produce outward or inward film polarization, respectively. At intermediate pO_2 values we observe a strong suppression of the Curie temperature. These experimental observations are qualitatively consistent with a thermodynamic model we have developed based on Landau-Ginzburg-Devonshire theory, which takes into account the chemical interaction between a gaseous environment and the polar surface of a ferroelectric.

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